

Attorney's Docket: 1999DE132  
Serial No.: 09/722,760  
Group: 1756

### AMENDMENTS TO THE CLAIMS

1) through 5) (Cancelled)

6) (Currently Amended) The method as claimed in claim 4 25, wherein R<sup>1</sup> to R<sup>18</sup> ~~denote is~~ hydrogen CN, CH<sub>2</sub>-CN, CF<sub>3</sub>, C<sub>1</sub>-C<sub>22</sub>-alkyl, C<sub>2</sub>-C<sub>18</sub>-alkenyl, C<sub>1</sub>-C<sub>18</sub>-alkoxy, C<sub>1</sub>-C<sub>18</sub>-hydroxy-alkyl,

C<sub>1</sub>-C<sub>18</sub>-halogenoalkyl, C<sub>2</sub>-C<sub>18</sub>-halogenoalkenyl, C<sub>1</sub>-C<sub>18</sub>-aminoalkyl, (C<sub>1</sub>-C<sub>8</sub>)-trialkylammonium-(C<sub>1</sub>-C<sub>18</sub>)-alkyl, (C<sub>1</sub>-C<sub>18</sub>)-alkylene-O(C=O)-(C<sub>1</sub>-C<sub>22</sub>)alkyl, (C<sub>1</sub>-C<sub>18</sub>)-alkylene-O(C=O)-phenyl, (C<sub>1</sub>-C<sub>18</sub>)-alkylene-NHCO-(C<sub>1</sub>-C<sub>22</sub>)alkyl, (C<sub>1</sub>-C<sub>18</sub>)-alkylene-NHCO-phenyl, (C<sub>1</sub>-C<sub>18</sub>)-alkylene-(C=O)O-(C<sub>1</sub>-C<sub>22</sub>)alkyl, (C<sub>1</sub>-C<sub>18</sub>)-alkylene-(C=O)O-phenyl, (C<sub>1</sub>-C<sub>18</sub>)alkylene-(C=O)NH-(C<sub>1</sub>-C<sub>22</sub>)alkyl, (C<sub>1</sub>-C<sub>18</sub>)-alkylene-CONH-phenyl, benzyl, phenyl, naphthyl, or C<sub>1</sub>-C<sub>12</sub>-alkylene-heterocyclyl;

R<sup>19</sup> ~~denotes is~~ C<sub>4</sub>-C<sub>5</sub>-alkylene, -(C<sub>2</sub>H<sub>4</sub>-O)<sub>1-9</sub>-(CH<sub>2</sub>)<sub>1-2</sub>- or -(C<sub>2</sub>H<sub>4</sub>-NH)<sub>1-9</sub>-(CH<sub>2</sub>)<sub>1-2</sub>;

R<sup>60</sup> ~~denotes is~~ C<sub>1</sub>-C<sub>18</sub>-acyl, C<sub>1</sub>-C<sub>18</sub>-alkyl, C<sub>2</sub>-C<sub>18</sub>-alkenyl, C<sub>1</sub>-C<sub>12</sub>-alkylene-phenyl, C<sub>1</sub>-C<sub>18</sub>-alkylene-pyridyl, phenyl or pyridyl;

R<sup>61</sup> and R<sup>64</sup> ~~denote are~~ -(CH<sub>2</sub>)<sub>1-12</sub>-, C<sub>1</sub>-C<sub>8</sub>-alkylene-phenylene, phenylene or C<sub>1</sub>-C<sub>8</sub>-alkylenepyridylene or piperidylene;

R<sup>71</sup> and R<sup>72</sup> ~~denote are~~ -(CH<sub>2</sub>)<sub>1-8</sub> and

R<sup>73</sup> and R<sup>74</sup> ~~denote are~~ hydrogen or (C<sub>1</sub>-C<sub>18</sub>)-alkyl.

7) (Currently Amended) A method of imparting, controlling or improving the charge of an electrophotographic toner or developer, or an electret material, consisting of adding only a structured silicate salt, wherein the cation is a low molecular weight, nonpolymeric ammonium ion or a combination of a low molecular weight, nonpolymeric ammonium ion with NH<sub>4</sub><sup>+</sup>, H<sub>3</sub>O<sup>+</sup>, an alkali metal, an alkaline earth metal, an earth metal or with a transition metal and the anion is an island, cyclic, group, chain, ribbon, laminar or matrix silicate or a combination thereof to a binder of an electrophotographic toner or developer or of an electret material, wherein the low

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molecular weight, nonpolymeric ammonium ion is ~~The method as claimed in claim 1,~~  
~~wherein the low molecular weight organic cation is an ammonium ion which is an~~  
~~aliphatic or aromatic 5- to 12-membered heterocyclic radical with 1 to 4 atoms~~  
~~selected from the group consisting of N, O, S and a combination thereof and S, or a~~  
~~combination thereof, belonging to the rings.~~

8) (Original) The method as claimed in claim 7, wherein the heterocyclic radical is pyridinium, pyridazinium, pyrimidinium, pyrazinium, purinium, tetraazaporphyrinium, piperidinium, morpholinium, tetrazonium, triaza-cyclononanium or tetraaza-cyclododecanium.

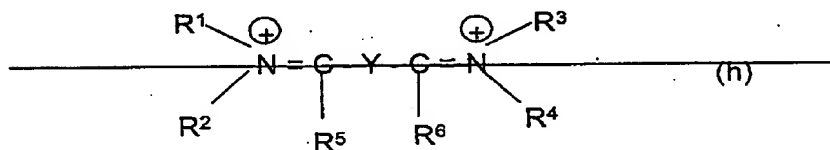
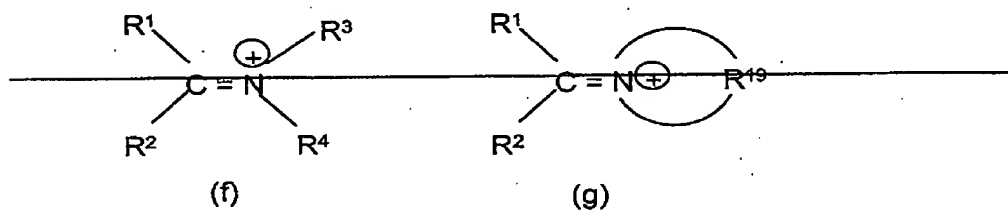
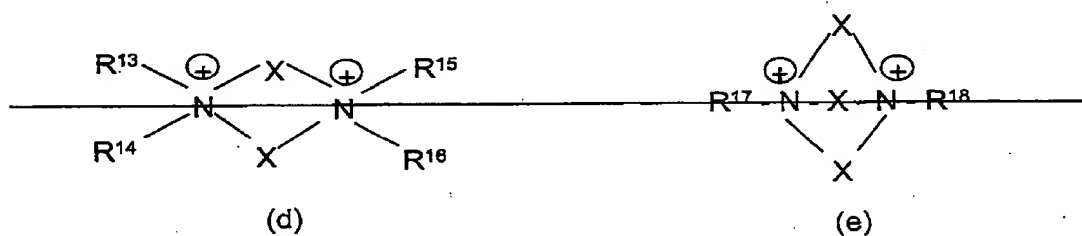
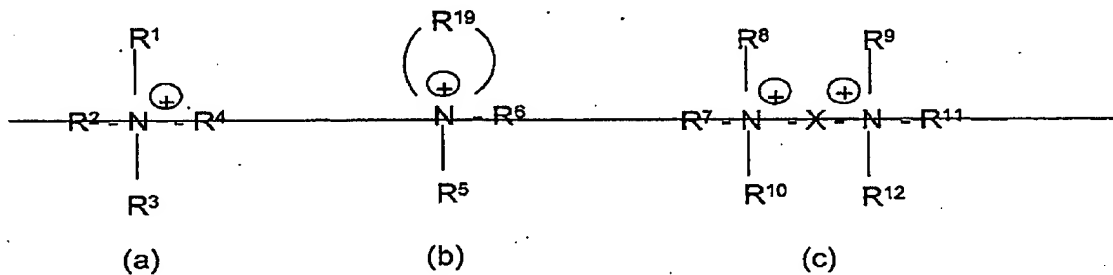
9) through 21) (Cancelled)

22) (Currently Amended) ~~A~~ The method of imparting, controlling or improving the charge of an electrophotographic toner or developer, or an electret material comprising the step of adding a charge control agent to a binder of an electrophotographic toner or developer or of an electret material, wherein the charge control agent according to claim 25, wherein the structured silicate salt is distearyl dimethyl ammonium bentonite.

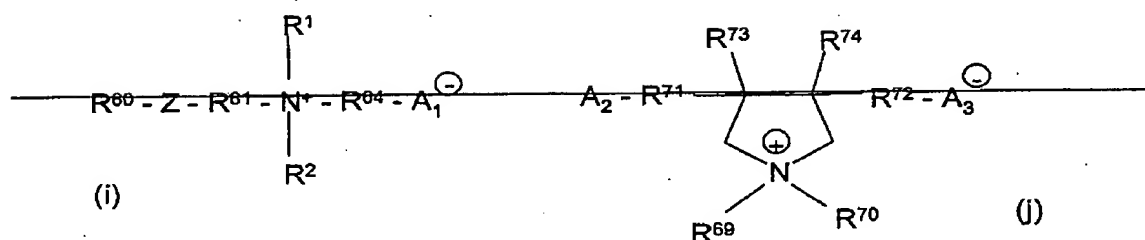
23) (Currently Amended) ~~A~~ The method according to claim 25, wherein the structured silicate salt of imparting, controlling or improving the charge of an electrophotographic toner or developer, or an electret material, comprising the step of adding as a charge control agent a structured silicate salt wherein the cation is a low molecular weight, nonpolymeric ammonium ion or a combination of a low molecular weight nonpolymeric ammonium ion with  $\text{NH}_4^+$ ,  $\text{H}_3\text{O}^+$ , an alkali metal, an alkaline earth metal, an earth metal or with a transition metal and the anion is an island, cyclic, group, chain, ribbon, laminar or matrix silicate or a combination thereof to a binder of an electrophotographic toner or developer or of an electret material, wherein the charge control agent imparts either a positive or negative charge;

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wherein the low molecular weight, nonpolymeric ammonium ion is of the formula (a) —(j)



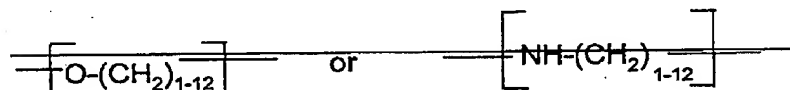
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in which

$R^1$  to  $R^{18}$  are identical or different and represent hydrogen, CN,  $(CH_2)_{4-18}$  CN, halogen, branched or unbranched  $C_4$ - $C_{22}$ -alkyl, mono- or polyunsaturated  $C_2$ - $C_{32}$ -alkenyl,  $C_4$ - $C_{22}$ -alkoxy,  $C_4$ - $C_{22}$ -hydroxyalkyl,  $C_4$ - $C_{22}$ -halogenoalkyl,  $C_2$ - $C_{22}$ -halogenoalkenyl,  $C_4$ - $C_{22}$ -aminoalkyl,  $(C_4$ - $C_{42})$  trialkyl ammonium  $(C_4$ - $C_{22})$ -alkyl,  $(C_4$ - $C_{22})$ -alkylene- $(C=O)O$ - $(C_4$ - $C_{32})$ -alkyl,  $(C_4$ - $C_{22})$ -alkylene- $(C=O)O$ -aryl,  $(C_4$ - $C_{22})$ -alkylene- $(C=O)NH$ - $(C_4$ - $C_{32})$ -alkyl,  $(C_4$ - $C_{22})$ -alkylene- $(C=O)NH$ -aryl,  $(C_4$ - $C_{22})$ -alkylene- $O(CO)$ - $(C_4$ - $C_{32})$ -alkyl,  $(C_4$ - $C_{22})$ -alkylene- $O(CO)$ -aryl,  $(C_4$ - $C_{22})$ -alkylene- $NH(C=O)$ - $(C_4$ - $C_{22})$ -alkyl, or  $(C_4$ - $C_{22})$ -alkylene- $NHCO$ -aryl,

wherein

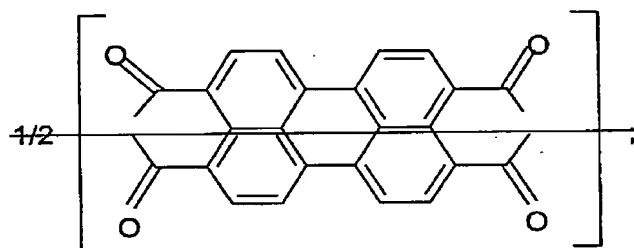
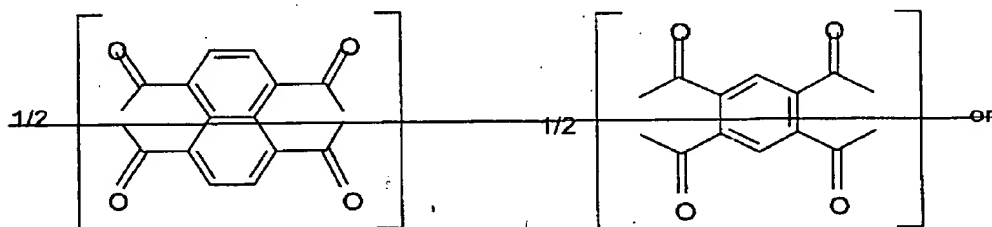
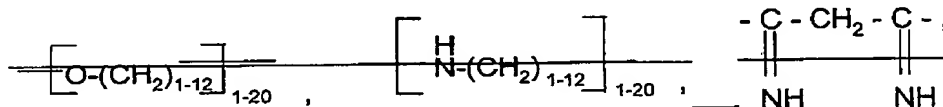


are optionally inserted into the acid ester or acid amide bonds;

$[(C_4$ - $C_{42})$ -alkylene- $O$ ]-H; aryl,  $(C_4$ - $C_{48})$ -alkylenearyl;  $(O-SiR'_2)-O-SiR'_3$ , in which  $R'$  has the meaning  $C_4$ - $C_{42}$ -alkyl, phenyl, benzyl or  $C_4$ - $C_{42}$ -alkoxy; heterocyclyl, or  $C_4$ - $C_{48}$ -alkylene-heterocyclyl, wherein the aryl and heterocyclyl radicals are optionally mono- or polysubstituted on carbon atoms or heteroatoms by  $C_4$ - $C_{42}$ -alkyl,  $C_4$ - $C_4$ -alkenyl,  $C_4$ - $C_4$ -alkoxy, hydroxy- $(C_4$ - $C_4)$ -alkyl, amino- $(C_4$ - $C_4)$ -alkyl,  $C_4$ - $C_4$ -alkylimino, carboxyl, hydroxyl, amino, nitro, cyano, halogen,  $C_4$ - $C_{42}$ -acyl,  $C_4$ - $C_4$ -halogenoalkyl,  $C_4$ - $C_4$ -alkylcarbonyl,  $C_4$ - $C_4$ -alkylcarbonyloxy,  $C_4$ - $C_4$ -alkoxy carbonyl,  $C_4$ - $C_4$ -alkylaminocarbonyl,  $C_4$ - $C_4$ -alkylcarbonylimino,  $C_6$ - $C_{40}$ -arylecarbonyl, aminocarbonyl, aminosulfonyl,  $C_4$ - $C_4$ -alkylaminosulfonyl, phenyl, naphthyl, or heteroaryl;

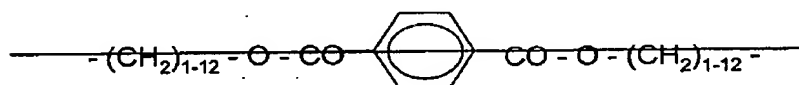
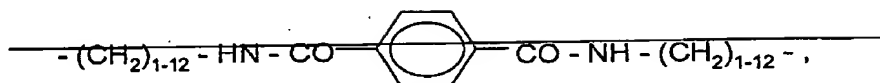
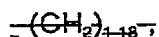
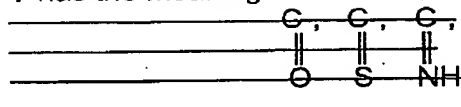
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$R^{40}$  represents  $C_4-C_{44}$ -alkylene,  $(C_2H_4-O)_{4-17}$ ,  $(CH_2)_{4-2}$  or  $(C_2H_4-NR)_{4-17}$ ,  $(CH_2)_{4-2}$ , in which R is hydrogen or  $C_1-C_{12}$ -alkyl;  
 X has the meaning of Y, or  $CO-CH_2-CO$ ,



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~~Y~~ has the meaning



or ~~o~~, ~~p~~, ~~m~~ ( $\text{C}_6\text{-C}_{44}$ ) arylene or ( $\text{C}_4\text{-C}_{44}$ ) heteroarylene with 1, 2, 3 or 4 heteroatoms selected from the group consisting of N, O, S and a combination thereof;

$\text{R}^{60}$  represents  $\text{C}_4\text{-C}_{32}$  acyl,  $\text{C}_4\text{-C}_{22}$  alkyl,  $\text{C}_2\text{-C}_{22}$  alkenyl,  $\text{C}_4\text{-C}_{48}$  alkylene  $\text{C}_6\text{-C}_{40}$  aryl,  $\text{C}_4\text{-C}_{22}$  alkylene heterocyclyl,  $\text{C}_6\text{-C}_{40}$  aryl or ( $\text{C}_4\text{-C}_{44}$ ) heteroaryl with 1, 2, 3 or 4 heteroatoms selected from the group consisting of N, O, S, and a combination thereof;

$\text{R}^{61}$  and  $\text{R}^{64}$  represent  $\text{-(CH}_2\text{)}_{4-18}$ ,  $\text{C}_4\text{-C}_{42}$  alkylene  $\text{C}_6\text{-C}_{40}$  arylene,  $\text{C}_6\text{-C}_{40}$  arylene, or  $\text{C}_6\text{-C}_{42}$  alkylene heterocyclyl;

Z represents ~~NH~~ or ~~O~~;

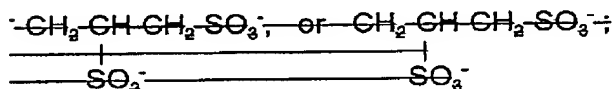
$\text{A}_4$  and  $\text{A}_5$  represent  ~~$\text{COO}^-$ ,  $\text{SO}_3^-$ ,  $\text{OSO}_3^-$ ,  $\text{SO}_3\text{H}$  or  $\text{CS}_2^-$~~ ;

$\text{A}_2$  represents  ~~$\text{SO}_2\text{Na}$ ,  $\text{SO}_3\text{Na}$ ,  $\text{SO}_2\text{H}$ ,  $\text{SO}_3\text{H}$  or hydrogen~~;

$\text{R}^{66}$  and  $\text{R}^{70}$  independently of one another represent hydrogen,  $\text{C}_4\text{-C}_{32}$  alkyl, in which the alkyl chain optionally contain one or more of the groups selected from the group

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consisting of ~~NH-CO~~, ~~CO-NH~~, ~~CO-O~~, and ~~O-CO~~; ~~C<sub>4</sub>-C<sub>48</sub>-alkylene-aryl~~, ~~C<sub>6</sub>-C<sub>48</sub>-alkylene-heterocyclyl~~, ~~C<sub>4</sub>-C<sub>48</sub>-hydroxyalkyl~~, ~~C<sub>4</sub>-C<sub>48</sub>-halogenoalkyl~~, ~~aryl~~, ~~(CH<sub>2</sub>)<sub>8</sub>-SO<sub>3</sub><sup>-</sup>~~;



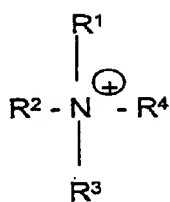
~~R<sup>71</sup> and R<sup>72</sup> represent (CH<sub>2</sub>)<sub>1-12</sub>; and~~

~~R<sup>73</sup> and R<sup>74</sup> represent hydrogen or C<sub>4</sub>-C<sub>22</sub>-alkyl.~~

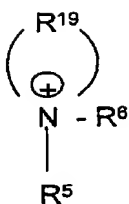
24. (Cancelled)

25. (Currently Amended) A method of imparting, controlling or improving the charge of an electrophotographic toner or developer, or an electret material, ~~comprising the step of adding as a charge control agent, wherein the charge control agent consists~~ consisting of adding only a structured silicate salt, wherein the cation is a low molecular weight, nonpolymeric ammonium ion or a combination of a low molecular weight, nonpolymeric ammonium ion with NH<sub>4</sub><sup>+</sup>, H<sub>3</sub>O<sup>+</sup>, an alkali metal, an alkaline earth metal, an earth metal or with a transition metal and the anion is an island, cyclic, group, chain, ribbon, laminar or matrix silicate or a combination thereof to a binder of an electrophotographic toner or developer or of an electret material, wherein the low molecular weight, nonpolymeric ammonium ion is ~~of the formula (a)~~ selected from one of the formulas (a) through (j)

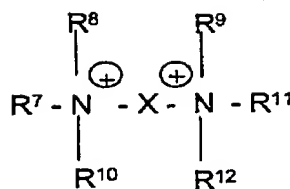
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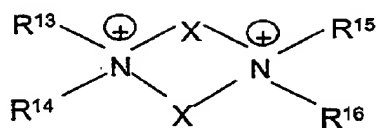
(a)



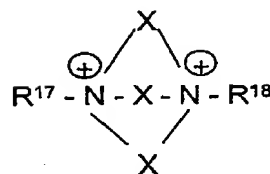
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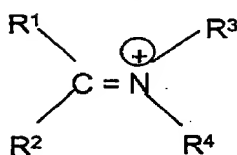
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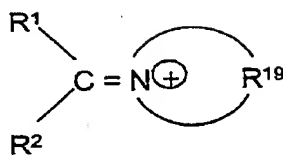
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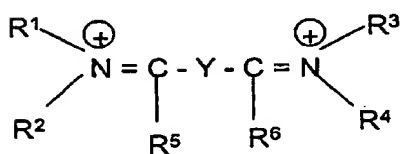
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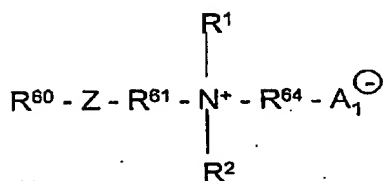
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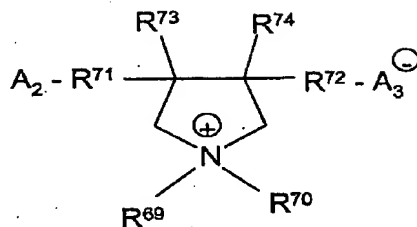
(g)



(h)



(i)



(j)

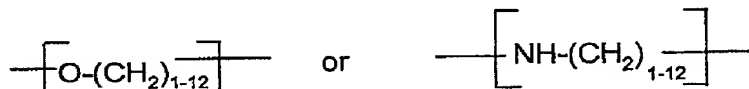


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in which

R<sup>1</sup> to R<sup>18</sup> are identical or different and represent hydrogen, CN, (CH<sub>2</sub>)<sub>1-18</sub>CN, halogen, branched or unbranched C<sub>1</sub>-C<sub>32</sub>-alkyl, mono- or polyunsaturated C<sub>2</sub>-C<sub>32</sub>-alkenyl, C<sub>1</sub>-C<sub>22</sub>-alkoxy, C<sub>1</sub>-C<sub>22</sub>-hydroxyalkyl, C<sub>1</sub>-C<sub>22</sub>-halogenoalkyl, C<sub>2</sub>-C<sub>22</sub>-halogenoalkenyl, C<sub>1</sub>-C<sub>22</sub>-aminoalkyl, (C<sub>1</sub>-C<sub>12</sub>)-trialkyl-ammonium-(C<sub>1</sub>-C<sub>22</sub>)-alkyl; (C<sub>1</sub>-C<sub>22</sub>)-alkylene-(C=O)O-(C<sub>1</sub>-C<sub>32</sub>)-alkyl, (C<sub>1</sub>-C<sub>22</sub>)-alkylene-(C=O)O-aryl, (C<sub>1</sub>-C<sub>22</sub>)-alkylene-(C=O)NH-(C<sub>1</sub>-C<sub>32</sub>)-alkyl, (C<sub>1</sub>-C<sub>22</sub>)-alkylene-(C=O)NH-aryl, (C<sub>1</sub>-C<sub>22</sub>)-alkylene-O(CO)-(C<sub>1</sub>-C<sub>32</sub>)-alkyl, (C<sub>1</sub>-C<sub>22</sub>)-alkylene-O(CO)-aryl, (C<sub>1</sub>-C<sub>22</sub>)-alkylene-NH(C=O)-(C<sub>1</sub>-C<sub>32</sub>)-alkyl, or (C<sub>1</sub>-C<sub>22</sub>)-alkylene-NHCO-aryl,

wherein



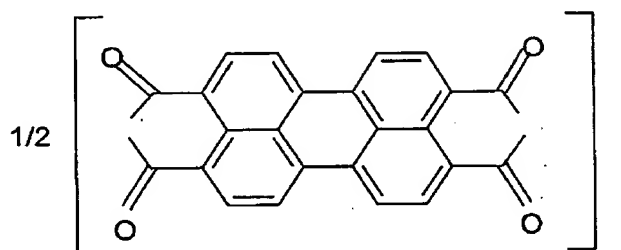
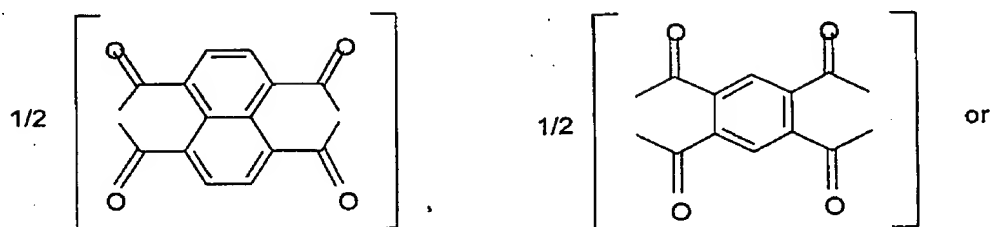
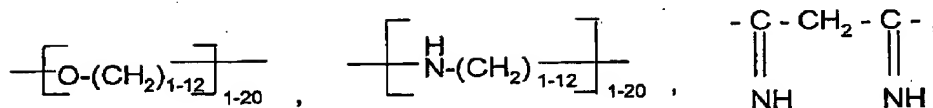
are optionally inserted into the acid ester or acid amide bonds;

[(C<sub>1</sub>-C<sub>12</sub>)-alkylene-O]-H; aryl, (C<sub>1</sub>-C<sub>18</sub>)-alkylenearyl; -(O-SiR'<sub>2</sub>)-O-SiR'<sub>3</sub>, in which R' has the meaning C<sub>1</sub>-C<sub>12</sub>-alkyl, phenyl, benzyl or C<sub>1</sub>-C<sub>12</sub>-alkoxy; heterocyclyl, or C<sub>1</sub>-C<sub>18</sub>-alkylene-heterocyclyl, wherein the aryl and heterocyclyl radicals are optionally mono- or polysubstituted on carbon atoms or heteroatoms by C<sub>1</sub>-C<sub>12</sub>-alkyl, C<sub>1</sub>-C<sub>4</sub>-alkenyl, C<sub>1</sub>-C<sub>4</sub>-alkoxy, hydroxy-(C<sub>1</sub>-C<sub>4</sub>)-alkyl, amino-(C<sub>1</sub>-C<sub>4</sub>)-alkyl, C<sub>1</sub>-C<sub>4</sub>-alkylimino, carboxyl, hydroxyl, amino, nitro, cyano, halogen, C<sub>1</sub>-C<sub>12</sub>-acyl, C<sub>1</sub>-C<sub>4</sub>-halogenoalkyl, C<sub>1</sub>-C<sub>4</sub>-alkylcarbonyl, C<sub>1</sub>-C<sub>4</sub>-alkylcarbonyloxy, C<sub>1</sub>-C<sub>4</sub>-alkoxycarbonyl, C<sub>1</sub>-C<sub>4</sub>-alkylaminocarbonyl, C<sub>1</sub>-C<sub>4</sub>-alkylcarbonylimino, C<sub>6</sub>-C<sub>10</sub>-arylcarbonyl, aminocarbonyl, aminosulfonyl, C<sub>1</sub>-C<sub>4</sub>-alkylaminosulfonyl, phenyl, naphthyl, or heteroaryl;

R<sup>19</sup> represents C<sub>4</sub>-C<sub>11</sub>-alkylene, -(C<sub>2</sub>H<sub>4</sub>-O)<sub>1-17</sub>-(CH<sub>2</sub>)<sub>1-2</sub>- or -(C<sub>2</sub>H<sub>4</sub>-NR-)<sub>1-17</sub>-(CH<sub>2</sub>)<sub>1-2</sub>-, in which R is hydrogen or C<sub>1</sub>-C<sub>12</sub>-alkyl;

X has the meaning of Y, or -CO-CH<sub>2</sub>-CO-,

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Y has the meaning  $\begin{array}{c} -\text{C}- \\ || \\ \text{O} \end{array}$ ,  $\begin{array}{c} -\text{C}- \\ || \\ \text{S} \end{array}$ ,  $\begin{array}{c} -\text{C}- \\ || \\ \text{NH} \end{array}$ ,  $-(\text{CH}_2)_{1-18}-$ ,



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or o-, p-, m-(C<sub>6</sub>-C<sub>14</sub>)-arylene or (C<sub>4</sub>-C<sub>14</sub>)-heteroarylene with 1, 2, 3 or 4 heteroatoms selected from the group consisting of N, O, S and a combination thereof;

R<sup>60</sup> represents C<sub>1</sub>-C<sub>32</sub>-acyl, C<sub>1</sub>-C<sub>22</sub>-alkyl, C<sub>2</sub>-C<sub>22</sub>-alkenyl, C<sub>1</sub>-C<sub>18</sub>-alkylene-C<sub>6</sub>-C<sub>10</sub>-aryl, C<sub>1</sub>-C<sub>22</sub>-alkylene-heterocyclyl, C<sub>6</sub>-C<sub>10</sub>-aryl or (C<sub>4</sub>-C<sub>14</sub>)-heteroaryl with 1, 2, 3 or 4 heteroatoms selected from the group consisting of N, O, S, and a combination thereof;

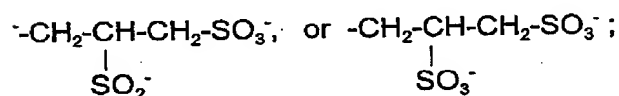
R<sup>61</sup> and R<sup>64</sup> represent -(CH<sub>2</sub>)<sub>1-18</sub>-, C<sub>1</sub>-C<sub>12</sub>-alkylene-C<sub>6</sub>-C<sub>10</sub>-arylene, C<sub>6</sub>-C<sub>10</sub>-arylene, or C<sub>0</sub>-C<sub>12</sub>-alkylene-heterocyclyl;

Z represents -NH- or -O-;

A<sub>1</sub> and A<sub>3</sub> represent -COO<sup>-</sup>, -SO<sub>3</sub><sup>-</sup>, -OSO<sub>3</sub><sup>-</sup>, -SO<sub>2</sub><sup>-</sup>, -COS<sup>-</sup> or -CS<sub>2</sub><sup>-</sup>;

A<sub>2</sub> represents -SO<sub>2</sub>Na, -SO<sub>3</sub>Na, -SO<sub>2</sub>H, -SO<sub>3</sub>H or hydrogen;

R<sup>66</sup> and R<sup>70</sup> independently of one another represent hydrogen, C<sub>1</sub>-C<sub>32</sub>-alkyl, in which the alkyl chain optionally contain one or more of the groups selected from the group consisting of -NH-CO-, -CO-NH-, -CO-O-, and -O-CO-; C<sub>1</sub>-C<sub>18</sub>-alkylene-aryl, C<sub>0</sub>-C<sub>18</sub>-alkylene-heterocyclyl, C<sub>1</sub>-C<sub>18</sub>-hydroxyalkyl, C<sub>1</sub>-C<sub>18</sub>-halogenoalkyl, aryl, -(CH<sub>2</sub>)<sub>3</sub>-SO<sub>3</sub><sup>-</sup>,



R<sup>71</sup> and R<sup>72</sup> represent -(CH<sub>2</sub>)<sub>1-12</sub>-; and

R<sup>73</sup> and R<sup>74</sup> represent hydrogen or C<sub>1</sub>-C<sub>22</sub>-alkyl.